A COMPREHENSIVE FRACTAL APPROACH IN DETERMINATION OF THE EFFECTIVE THERMAL CONDUCTIVITY OF GAS DIFFUSION LAYERS IN POLYMER ELECTROLYTE MEMBRANE FUEL CELLS

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Special dedication to my beloved mother and father
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ABSTRACT

The challenges in the fuel cell industry is to produce the efficient thermal and water management for accurate determination of the effectiveness thermal conductivity of gas diffusion layers (GDL) used in polymer electrolyte membrane fuel cells (PEMFC's). This is one of the factors affecting the durability of a fuel cell and need to get a solution to minimize costs and optimize the use of electrodes and cells. The main objectives of this research focus on the capability of the fractal approach for estimation the effectiveness of thermal conductivity of gas diffusion layer. Moreover, on this research also to propose modified fractal equations in determination of the effective thermal conductivity of GDL in PEMFCs based on previous study. Other objectives in this study are demonstrated the thermal conductivity of GDL treated with PTFE contents by using through-plane thermal conductivity experiment method. The through-plane measurement (experiment method) has been used in estimating through-plane thermal conductivity of the GDL. Thermal resistance for GDL also has been investigated under compression pressure 0.1 MPa until 1.0 MPa. In fractal equation, the determination of tortuous and pore fractal dimension can be done by using Scanning Electron Microscopy (SEM) method. Determination of effectiveness thermal conductivity using of fractal equation with slightly modified. In findings, it was found that fractal equation have been modified and measured on the GDL parameter characteristics. It was shown that the value of the effectiveness thermal conductivity of the sample using fractal approach is in good agreement with the experimental value. Finally, all the effective thermal conductivity measured by experimental and fractal approach have been determined with the variant temperature and compression pressure to show the validation result between of this two methods.

ABSTRAK

Cabaran dalam industri *fuel cell* adalah untuk pengurusan haba dan air yang cekap dan juga penentuan tepat secara keberkesanan bagi keberaliran haba pada lapisan resapan gas (GDL) yang digunakan dalam membran elektrolit polimer *fuel cell* (PEMFCs). Ini adalah merupakan salah faktor-faktor yang mempengaruhi ketahanan pada *fuel cell* dan hal ini perlu mendapat penyelesaian yang terbaik untuk mengurangkan kos dan mengoptimumkan penggunaan elektrod dan sel-sel. Objektif utama fokus penyelidikan ini adalah untuk mengetahui keupayaan pendekatan fraktal untuk anggaran keberkesanan kekonduksian haba pada PEM dan GDL. Selain itu, kajian ini juga untuk mencadangkan pengubahsuaian terhadap persamaan fraktal dalam penentuan kekonduksian haba yang berkesan GDL di PEMFCs berdasarkan kajian sebelumnya. Objektif lain dalam kajian ini menunjukkan keberaliran haba pada GDL dengan kandungan PTFE berbeza dengan menggunakan kaedah eksperimen kekonduksian haba melalui ujian pelantar. Pengukuran ujian pelantar (kaedah eksperimen) telah digunakan dalam menganggarkan kekonduksian haba daripada GDL. Rintangan haba untuk GDL juga telah dijalankan di bawah tekanan mampatan 0.1 sehingga 1.0 MPa. Dalam persamaan fraktal, penentuan kelikuan dan liang dimensi fraktal boleh dilakukan dengan menggunakan kaedah *Scanning Electron Microscopy* (SEM). Penentuan keberkesanan kekonduksian haba menggunakan persamaan fraktal dengan sedikit pengubahsuaian telah dijalankan. Hasilnya telah didapati bahawa nilai keberkesanan keberaliran haba terhadap bahan ujikaji dengan menggunakan kaedah pendekatan fraktal adalah bersamaan dengan nilai yang diperoleh dengan menggunakan kaedah eksperimen. Akhir sekali, kesemua nilai keberkesanan kadar aliran haba telah diukur dengan kaedah eksperimen dan fractal telah ditunjukkan debgan berlainan suhu pemanasan dan tekanan mampatan yang berbeza bagi menunjukkan pengesahan keputusan bagi kedua-dua kaedah ini.

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LIST OF SYMBOLS AND ABBREVIATIONS

ΔT	-	Temperature drop across thin film
ξ	-	Ratio of the number of perpendicular channels to the total number of channels
λ	-	Size of the path
λ_{min}, λ_{max}	-	The minimum and maximum pore diameters
τ	-	Tortuous pathways
$\mathcal{E}$, $\emptyset$	-	Porosity
A	-	Total area of a structural size
A_s	-	Area of heat transfer
C	-	Intercept of the line constant to the data in a log-log plot
C_{ph}	-	Phonon heat capacity
DAQ	-	Data Acquisition
D_f	-	Fractal dimension
D_p	-	Specific pore fractal dimension
D_t	-	Tortuous fractal dimension
$f(\xi)$	-	The effects of anisotropy and structure in the in-plane and through plane directions
FC	-	Fuel Cell
GDL, DM	-	Gas Diffusion Layer and Diffusion Media
k	-	Thermal conductivity
k_g	-	Thermal conductivity of the occupied gas
k_{s1}	-	Thermal conductivity of the carbon fiber
k_{s2}	-	Thermal conductivity of the PTFE
K_{eff}	-	Effective thermal conductivity
$K_{eff,s}$	-	Effective thermal conductivity of solid phase

$K_{eff,p}$	-	Effective thermal conductivity of gas phase
k_{mem}	-	Thermal conductivity of PEM and GDL
$L(\lambda)$	-	Corresponding path of length
L_o	-	Linear length of capillary pathway in the flow direction
L_s	-	Thickness of PEM and GDL
$M(L)$	-	Fractal structure
MEA	-	Membrane Electrode Assembly
MPL	-	Microporous layer
N	-	The cumulative pore size distribution
P	-	Pressure
PEM	-	Polymer Eletrolyte Membrane
PEMFCs	-	Polymer Eletrolyte Membrane Fuel Cells
PTFE	-	Polytetrafluoroethylene
Q	-	Heat flow rate
R	-	Pore size
r	-	Pore radius
R	-	Thermal resistance
$R_{H\text{-}sample}$	-	Contact resistance between the sample and the holder
R_L	-	Lower resistance
r_{max}	-	Maximum pore radius
R_{sample}	-	Thermal of sample
R_{total}	-	Total thermal resistance
R_U	-	Upper resistance
SEM	-	Scanning Electron Microscope
T	-	Temperature
T_c	-	Temperature of the cold plate
T_h	-	Temperature of the hot plate
V_p	-	Pore volume related with a certain pressure
v_{ph}	-	phonon velocity
I_{ph}	-	Phonon mean free path

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CHAPTER 1

INTRODUCTION

1.1 Research background

The renewal and cleanest energy sources become more serious attention in nowadays due to the depletion of petroleum resources and potential to reduce emission pollution. In current high demand on that, the one of the most efficient environmental-friendly generation of energy is called fuel cell technology. Fuel Cell technology have received more attention in recent years is due to the high efficiency and low emissions. There are several categories of fuel cell which is Polymer Electrolyte Membrane (PEM) fuel cells or PEMFC, Solid Oxide Fuel Cells (SOFC), Phosphoric Acid Fuel Cells (PAFC), Alkaline Fuel Cells (AFC) and Molten Carbonate Fuel Cells (MCFC). The focusing on this research is Polymer electrolyte membrane fuel cells (PEMFC), where constructed from membrane electrode assembly (MEA) including electrodes, electrolytes, catalysts, and gas diffusion layers. The polytetrafluoroethylene (PTFE) Nafion® membrane acting as proton conductor and (Pt)-based material as catalyst. PEMFCs convert chemical energy stored in hydrogen (as a fuel) and oxygen directly and efficiently into electrical energy with water as the only byproduct, have the ability to reduce energy consumption, pollutant emissions and dependence to generation of electric energy using fossil fuels. Figure 1.1 Efficiency of PEMFC can reach as high as 60 % in the overall conversion of electrical energy and 80 % in cogeneration of electricity and heat with a reduction of more than 90 % in the primary pollutants (in United State of America).

The main applications of PEMFCs not only focusing on transportation, but it does include of portable and stationary power generation. In automotive industries, the well-known company as Honda, Toyota, General Motor and Hyundai has been developed and demonstrated their product based on technology fuel cell (Fuel Cell Vehicle) not only to fuel energy consumption saving but because of the potential impact on the environment, such as emissions control greenhouse gases (Wang *et al.*, 2011) (Wu *et al.*, 2014) (You *et al.*, 2017).

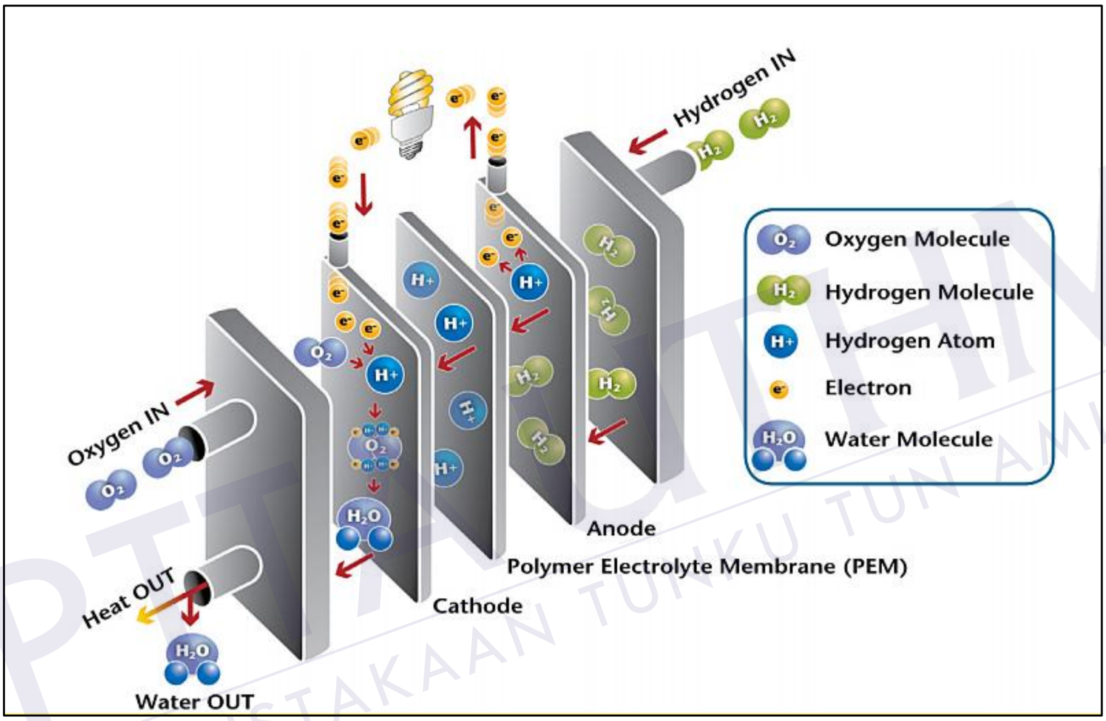

Figure 1.1: A functionally description of fuel cells (Energy Design Resources, 2013)

Since PEMFCs is a renewable energy source of the cleanest, and fuel (hydrogen and oxygen) are abundant in the earth, there are many researchers are racing to make fuel cells are much more efficient and low cost. Recently, considerable progress has been in improving power density, stability operations and design structure PEMFCs. However, there are still many challenges to overcome for profitable commercial applications. The challenges in fuel cell are its durability of life time while operating. Among the factors that predispose to failure of PEMFCs is the membrane manufacturing process, the material properties of the cell components, installation of the fuel cell, and fuel cell system operating conditions. In application fuel cell in transport, the frequency starting up and shutting down or nearly power random cycle load are effect the humidity condition of Nafion membrane and also to GDL. (Wu *et al.*, 2014)

The main challenge in the design of fuel cells is to transfer heat from the gas diffusion layer (GDL) in the polymer electrolyte membrane due to the result of a chemical reaction to produce electricity. Analysis of this process requires the determination of the effective thermal conductivity and thermal contact resistance is also associated with the interface between the GDL and the adjacent surface or coating. However, thermal conductivity of diffusion media or GDL is more difficult to estimate due it porosity structure. GDL porosity makes it necessary to use effective thermal conductivity to describe heat transfer in solid and liquid phases. Because of GDL is anisotropic and having high porosities, there are widely dispersed in literature thermal conductivity values.

In measuring thermal conductivity, there are several methods can be used. The prediction methods for effective thermal conductivity of porous media can be predicted by empirical formulas, numerical simulations, or theoretical models. The empirical formulas and numerical simulation have several issues where empirical method only based on simplifying assumption and empirical constants didn't indicate any specific physical meanings. For numerical simulation method, the difficult to analyst detail of GDL geometry model of porosity structure. (Nikooee *et al.*, 2011) Fractal approach can be used to estimate the thermal conductivity of diffusion media. Fractal theory can analyze complex structure such as high porosities characteristic. Fractal theory widely used in diverse engineering application which is involve physical phenomena in disordered structures and over multiple scales.

1.2 Problem statement

The main objectives for all researchers in the development of novel membrane materials are to improve the performance and durability of the fuel cells and reduce the overall cost of fuel cells. Thermal conductivity of the PEM components must be estimated accurately for better understanding the process of heat transfer in proton exchange membrane fuel cells. The electrochemical reaction and irreversibility associated in proton exchange membrane fuel cells generate large amounts of heat which produces a temperature gradient in the various components of the cell.

In literary studies, Nafion commonly used as the thermal conductivity of the membrane. GDL is one of the main components in MEA. The center part in MEA is Proton Exchange Membrane or Polymer Electrolyte Membrane (PEM). Figure 1.2 shows the schematic of MEA and their components. However, it is difficult to estimate the thermal conductivity of thermal diffusion media or gas diffusion layer because of GDL is anisotropic and having high porosities. However GDL porosity makes it necessary to use an effective thermal conductivity to describe heat transfer in solid and liquid phases. As mentioned before, in having high porosity materials and also anisotropic of GDL, which is probably be the reason why the thermal conductivity are widely scattered in the literature (Zakil *et al.*, 2016).

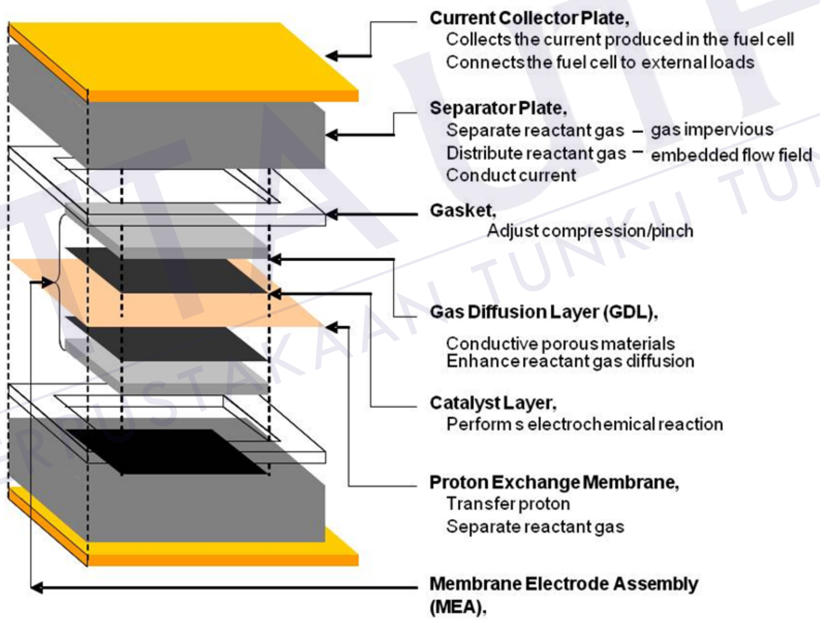

Figure 1.2: Schematic of Membrane Electrode Assembly (MEA) in PEMFCs (Iranzo, 2017)

Among the methods used including of empirical formulas, numerical simulations, or theoretical models to in determine the thermal conductivity, it's experiencing one of the following three things: analytical methods are usually based on very simplifying assumptions. Numerical solution with realistic assumptions that can capture details GDL as heterogeneous porous media usually takes time using simulations. The Empirical relationships including significant physical constants. In that case, fractal methods have been recommended by Nikoee *et al.*, (2011) as the successful method to estimate the thermal conductivity of diffusion media.

Fractal geometry has been widely used in recent years to characterize the complex, heterogeneous porous media. The main idea behind fractal geometry is the scale parameter extraction invariant, which can describe the structure of complex geometry. Physics phenomena that occur in such media can be associated with this parameter. The main role of fractal geometry, in this case, is to simplify the structure of porous media complex diffusion into the fractal dimension and the physical basis for the derivation of the targeted phenomena.

There are different fractal dimensions that describe the characteristics of the pores in the porous medium. Among the various fractal dimension, fractal dimension and tortuous pore fractal dimension (called pores or areas based on the number of fractal dimension calculation method) is the most important. Parameters that can be measured by simple experiment can be used to develop the thermal conductivity equation. To overcome these weaknesses, fractal geometry will be proposed as an approach that will help to estimate the effective thermal conductivity of the polymer electrolyte membrane.

1.3 Significance

This study aim to contributed regarding the temperature and heat transfer mechanism to determine the thermal conductivity of the components of the membrane electrode assembly and gas diffusion layer in order to approach of fractal method.

1.4 Objectives

This study embarks on the following objectives:

i. To investigate the capability of fractal approach in order to determine thermal conductivity in GDL and propose a new approach (fractal equation) in determination of the effective thermal conductivity of GDL in polymer electrolyte membrane fuel cells.

ii. To validate by varying temperature, pressure and thermal conductivity of GDL by using experimental method (through-plane method).

1.5 Scopes of study

The scopes of this study are listed as below:

i. An experimental study to determine the thermal conductivity (experimental) of the PEM samples; coated Nafion® 117 and untreated Nafion 117, the GDL samples; CT, ELAT® LT1400W, Sigracet 35 AA (0 % PTFE) and Sigracet® 35 BA (50 % PTFE) and additional of thermal contact resistance experiment between PEM and GDL samples with a metal plate as a function of temperature and pressure.

ii. Comparisons have been made between the fractal methods and the existing experimental results measured by one dimensional through-plane thermal conductivity.

iii. Using equation of effective thermal conductivity in series and parallel model to obtain pore fractal dimension from GDL pore size distribution.

iv. Scanning Electron Microscope (SEM) images of diffusion media also had been used, and conventional methods of fractal image processing such as box counting can be used to determine the pore fractal dimension.

v. The effect of the temperature on the through-plane thermal conductivity of all the components in the PEM and GDL was investigated in the temperature range 27 ºC - 90 ºC.

vi. The measurements temperature along the standard material and specimens were performed under camber in order to eliminate the heat transfer by convection.

vii. The compression pressure ranges between of 0.1 to 1 MPa.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In Polymer Electrolyte Membrane Fuel Cell (PEMFC) the membrane and catalyst layer (consisting of the MEA), both of which require further research is essential to identify and develop cost-effective material alternatives. Correlation properties of the membrane for the performance of polymer electrolyte materials generally are more in need. The process of heat transfer in porous GDL has been investigated and appropriate predictive model of effective thermal conductivity by using fractal theoretical characterization of the actual microstructure of GDL (Wang *et al.*, 2011).

In general, the heat transfer mechanism is controlled by convection, radiation and conduction. When the flow of liquid happen in pores the convection heat transfer were occurs. Generally, the effect of convection is more pronounced in the case of large pore size, while it is negligible for small pore size less than 100 µm at a lower temperature (below 373 K) due to lack of intensive fluid- circulation in the pores. In PEMFC, the GDL operating temperature produce in system is below than 375 K due to its pore sizes which is 100 µm and in this scenario the convection or radiation can neglected. (Shi *et al.*, 2008) (Kantorovich *et al.*, 1999).

Radiation heat transfer occurs through the release or absorption of heat radiation pore walls. From studies on the type of coal, it has been found that radiation mechanism has a significant effect on heat transfer through large pore size (> 10 μm) at high temperatures (>1000 K), while foremost carbon material, the effect of radiation heat transfer is negligible for temperatures below 1000 K (Shi *et al.*, 2008). Table 2.1 show summarizes properties of two type GDL materials. According to table, its show that the pore size of GDL is less than 100 μm (Wang *et al.*, 2016).

Table 2.1: Properties of two type GDL material (Wang *et al.*, 2016)

Material	PTFE loading (%)	Porosity	Pore size (μm)
sssCarbon cloth	0	0.78	97
Carbon paper TGP-H-120	0	0.81	32
Carbon paper TGP-H-120	20	0.74	32
Carbon paper TGP-H-120	70	0.61	31
Carbon paper TGP-H-120	97	0.57	32
Carbon paper TGP-H-120	120	0.52	30
Carbon paper TGP-H-120	150	0.43	32

2.2 Application of fuel cell

Technologies of fuel cells have got an attention in the transportation industry including public transport and private vehicles. This is because the potential energy produced by the fuel cell is capable of producing high efficiency, economical used and the ability to reduce the environmental pollution caused by carbon monoxide gas (Wang *et al.*, 2011).

System Proton exchange membrane fuel cells operating on hydrogen and has a power density as high as 1:35 kW / liter have been shown. This system has been integrated into the vehicle concept by a number of manufacturers. Specific design features of the fuel cell system 60 kW and operate on hydrogen supplied from a cryogenic liquid hydrogen tank. This vehicle has a range of 400 km, a top speed of 135 km/h, and can be accelerate 0 - 100 km/h in less than 16 s (Ellis *et al.*, 2001).

Development of fuel cell technology not only focusing on transportation, but there are many of manufacturer working on other application for example, vending machine, vacuum cleaners machines and traffic light. The growing of fuel cell technologies also involved of power generation for hospital, police station and the bank because of the advantages its-self (Segura *et al.*, 2009).

2.3 Proton exchange membrane fuel cell

There are many types of fuel cell as explained in Chapter 1. As PEMFC were selected, by using proton (hydrogen ion) conducting membrane will remain squeezed between two porous platinum-catalyzed electrodes. At first, this membrane is based on polystyrene, but currently Teflon-based products namely "Nafion" is used. It offers high stability, high oxygen solubility, good thermal and high mechanical stability. Nafion membrane is widely used for PEMFCs and have variety thickness and specific application. These membrane are proton-conductive polymer film, also as known as electrolyte or ionomer. The membrane functions are to allow only protons to through-pass to cathode (Oxygen) from anode (Hydrogen) as shown in Figure 2.1. Others are to spate the anode and cathode compartment of PEMFCs. PEMFCs are low temperature fuel cell and operating temperature is relatively low (between 60 – 100 °C). Hydrogen fuel cells require oxygen for humidified and operations. Pressure, in general, remains the same on both sides of the membrane. Operating at high pressure is necessary to achieve high power density, especially when the air is selected as the anodic reactants. The PEMFC meets the demands of rapid startup, acceleration, high power density and reliability. PEMFC will be best for light-duty vehicle applications (T-raissi *et al.*, 1992) (Mekhilef *et al.*, 2012).

As known the mechanical strength very important for membrane since the activities of the proton conductivity of the membrane. Figure 2.1 shows the illustration movement of hydrogen proton cross-over the membrane. Membranes, for example, need to carry protons, but not electrons. It is as thin as possible, so that proton current is affected as little as possible and the voltage drop across the

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